

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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Title	: BIAXIALLY ORIENTED WHITE	
	: POLYPROPYLENE FILM FOR	
	: THERMAL TRANSFER RECORDING	
	: AND RECEIVING SHEET FOR	
	: THERMAL TRANSFER RECORDING	
	: THEREFROM	

DECLARATION OF MR. RYOSUKE MATSUI

Commissioner for Patents
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Sir:

I, Ryosuke Matsui, declare that:

I reside at Kyoto, Japan

I am thoroughly familiar with the above identified patent application and the subject matter described and claimed therein;

I have a Master's degree in engineering which was conferred upon me by the Graduate School of Kobe University in 1996. I majored in Polymer Science in graduate school. In 1996, I began employment with Toray Industries, Inc. Since then I have been a researcher in Toray's Films & Film Products Research Laboratories.

I am now a Senior Research Chemist and perform research and development of biaxial stretched polyester films for packaging and molding applications and micro-porous polyolefin films. Therefore, I am highly skilled in film-making processes (consisting of film extrusion,

stretching and heat treatment).

I am familiar with the Official Action dated November 27, 2009 wherein Sadamitsu is used in combination with Asakura to reject all of the claims in this application. The rejection utilizes Sadamitsu for the teaching of the claimed term "cushion factor," that it would be obvious to optimize that cushion factor and that it would be reasonable to believe that the films of Sadamitsu would have a cushion factor within the claimed range.

I conducted experiments which include one example of a film produced in accordance with the teachings of this application and Claim 1. I also produced a film in accordance with the teachings of Sadamitsu and then conducted a series of tests on the resulting films. Details of the experiments follow and a table comparing the results follows the detailed description.

This application - Example 1

A homopolypropylene resin 99.9% by weight (hereafter, referred to as H-PP) (produced by Mitsui Chemicals, Inc., MFR: 4 g/10 min, I1: 98.5%) and N,N'-dicyclohexyl-2,6-naphthalene dicarboxamide (NU-100 produced by New Japan Chemical Co., Ltd.), 0.1% by weight as β -crystal nucleating agent were mixed and supplied to a twin screw extruder to thereby be melted and mixed at 280°C. The mixture was extruded, cooled by passing through a water bath of 20°C and cut by a chip cutter into 3 mm lengths, and then dried at 100°C for 2 hours. The β -crystal ratio of the β -crystal nucleating agent added PP (hereafter, abbreviated as β -crystal PP) was 82%.

Next, the β -crystal PP was fed to an extruder heated at 200°C and melted, extruded in a sheet through a monolayer T-die, closely contacted with a metal drum (casting drum) heated to a surface temperature of 90°C, cooled and solidified by blasting 30°C with cold air from the non-drum side and an undrawn film was produced. The contact time with the metal drum was 35

seconds.

Next, after the undrawn film was preheated by introducing to an oven heated and kept at 120°C, the film was drawn 4.5 times in length direction (longitudinal direction, namely, running direction of the film, hereafter it is abbreviated as MD direction), and cooled with a roll of 100°C. Then, the film drawn in the MD direction was introduced in a tenter by grasping both ends of the film with clips and drawn 10 times in the direction perpendicular to the MD direction (transverse direction, hereafter, abbreviated as TD direction) (areal drawn ratio: longitudinal draw ratio x transverse draw ratios=45 times) in an atmosphere heated to 135°C. To complete the crystal orientation of the biaxially oriented white polypropylene film and thereby impart smoothness and dimensional stability, relaxation heat treatment of 5% in the transverse direction was successively performed at 150°C in a tenter, and, after cooling slowly and uniformly, cooled to room temperature. Corona discharge treatment on both sides was performed in air to thereby make the wet tension into 37 mN/m and wound to provide surfaces to receive coatings or other substrates.

The thickness of the film thus obtained was 35µm, and by SEM observation of a film cross section, it was confirmed that the film contains many fine and non-nucleus voids inside. Next, after a paper of 150 µm thickness was pasted to the D side of the white film, the above-mentioned coating liquid for forming a receiving layer was coated with a micro gravure coater on the opposite surface side (ND side) of the D side which has a high glossiness, such that the coated amount was 3 g/m² when dried, and thereby obtained a receiving sheet for thermal transfer recording.

Sadamitsu - Example 1

N,N'-Dicyclohexyl-2,6-naphthalenedicarboxamide (0.2 weight part, used as a β-crystal nucleating agent) and 0.05 weight part of Irgafos 168 and 0.05 weight part of Irganox 1010 made

by Ciba Specialty Chemicals (used as antioxidants) were mixed in a Henschel mixer with 100 weight parts of a propylene-ethylene block copolymer with an MFR of 2.7 g/10 minutes and an ethylene content of 6.2 wt %. This mixture was melt mixed at 240°C in a single screw extruder, and the extruded resin was cooled and cut to prepare resin pellets containing the β -crystal nucleating agent.

The resin pellets were then extruded in the form of a sheet at a resin temperature of 220°C using a T-die extruder (twin screw extruder with a screw diameter of 65 mm, plus a T-die with a width of 350 mm). The sheet was cooled and solidified by being placed for 12 seconds on a chill roll with a diameter of 600 mm and maintained at a surface temperature of 120°C, resulting in an unstretched polypropylene web sheet with a width of 300 mm and a thickness of 380 μm .

The sheet was then guided to a longitudinal stretching apparatus with a roll surface temperature of 90°C, where it was stretched longitudinally at a ratio of 4 times, resulting in longitudinally stretched sheet with a width of 165 mm. The distance between the longitudinal stretching rolls was 435 mm, and the neck-in ratio in the width direction of the unstretched web sheet was 45%.

The longitudinally stretched sheet was then annealed while being longitudinally stretched at a stretch ratio of 10% with a roll having a surface temperature of 145°C. The annealing contact time during which the longitudinally stretched sheet was contacted with the roll was 5 seconds.

The annealed sheet was then guided to a transverse stretching apparatus, where it was subjected to transverse tenter stretching at a ratio of 6.0 times at a temperature of 140°C and a strain rate of 100%/sec, whereby a white, translucent stretched film was continuously obtained.

Measurement Method - Sadamitsu

Porosity

The stretched film was cut into a square and the length of one side (L cm), the weight (W g), and the thickness (D cm) were measured, and the porosity was calculated from the following equation.

$$\text{Porosity} = 100 - 100(W/p)/(L^2 \times D)$$

wherein p is the density of the unstretched polypropylene web sheet prior to stretching.

Pore Size

The pore size was determined by the bubble point method (JIS K 3832), by mercury intrusion porosimetry, and by electron microscope (SEM) observation of a film cross section.

Bubble point (BP) method: The average pore size and maximum pore size were measured using a bubble point type pore size measurement apparatus ("Permoporometer CFP-1200AEL" made by PMI).

Mercury intrusion porosimetry: Assuming that the pores were cylindrical, their pore size was calculated from the following equation using the total pore volume (V) and the pore specific surface area (A) obtained from a mercury intrusion porosimetric pore size measurement apparatus (Micromeritics AutoPore III model 9420, made by Shimadzu Seisakusho).

$$\text{Average pore size} = 4V/A$$

SEM observation: A porous film that had been cut to a size of 3 cm square was immersed in molten paraffin at 70°C, and the film was impregnated with the paraffin until the film became semitransparent. Then, the film was taken out and the paraffin was cooled and solidified. The film was then thoroughly cooled by bringing it into close contact with dry ice, and the film was cut with a razor blade in the longitudinal and transverse directions of the film. The impregnating paraffin was then removed by extraction with hexane, and the film was dried. Gold was deposited with an ion sputtering apparatus (Ion Sputter JFC-1100 made by JEOL) to

produce a film cross section observation sample. This was placed under an electron microscope (JSM-T200 made by JEOL), and micrographs were made of the film cross section at a magnification of 1000 times to obtain cross-sectional images including the film surface. The maximum pore size in the transverse direction, longitudinal direction, and thickness direction were read from cross-sectional images in the transverse and longitudinal directions.

This application - Cushion Factor

A dial gage type thickness meter (JIS B 7503 (1997), UPRIGHT DIAL GAUGE (0.001.times.2 mm) No. 25 produced by PEACOCK, gage head 5 mm ø flat type) is equipped with a dial gage stand (No. 7001 DGS-M). The film thickness obtainable by this (d0) is measured. Furthermore, the thickness when 500 gf load is applied to a dial gage press element (d500) is measured, and the cushion factor was calculated by the following formula (unit: %).

$$\text{Cushion factor (\%)} = \{(d0 - d500) / d0\} \times 100.$$

The same measurement was carried out 5 times for the same sample, and the average of the obtained cushion factor was defined as the cushion factor of said sample.

Results

Table

		Example 1 This application	Example 1 Sadamitsu
Average pore size (μm)	BP method	0.04	0.05
	Mercury intrusion method	0.18	0.25
Maximum pore size (μm)	BP method	0.04	0.055
	SEM observation /longitudinal direction	5.3	22
	/transverse direction	5.5	24
	/thickness direction	0.2	4.0
Porosity (%)		50	58
Cushion Factor (%)		23	4(<16)

Reference to the above table shows that the cushion factor between the film produced in accordance with this application was 23 which is within the middle of the about 16 to about 30 range specified in Claim 1. In sharp contrast, the cushion factor in Sadamitsu was 4%. This cushion factor is sharply outside of the lowest end of the Applicants' claimed range of about 16%. I therefore believe that the cushion factor produced by the Applicants in this application is quite unexpected based on the teachings of Sadamitsu.

The undersigned declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and thus such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: Jan. 20, 2010

Ryosuke Matsui
Ryosuke Matsui, Co-inventor